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Editorial

Mass Spectrometry Imaging in ACS Journals

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Mass spectrometry imaging (MSI) has rapidly evolved into a powerful and versatile analytical platform, enabling spatially resolved molecular characterization across diverse chemical, biological, and biomedical systems. This ACS Editors' Collection highlights recent advances in MSI that span methodological innovation, integration with complementary technologies, and impactful applications, reflecting the breadth and vitality of the field. Contributions curated from *ACS Measurement Science Au*, *Analytical Chemistry*, *Chemical & Biomedical Imaging*, *Journal of the American Society for Mass Spectrometry*, and *Journal of Proteome Research* showcase how advances in instrumentation, sample preparation, data analysis, and computational approaches continue to expand the capabilities and reach of MSI. Together, these articles provide a cross-journal perspective on emerging directions in mass spectrometry imaging and underscore its central role in modern measurement science, spatial omics, and chemical and biomedical imaging.

ACS MEASUREMENT SCIENCE AU

ACS Measurement Science Au focuses on new innovations in all areas of measurement science across separations, mass spectrometry, spectroscopy, electrochemistry, and imaging. We are especially interested in emerging multidisciplinary areas, and mass spectrometry imaging is at the forefront of multidisciplinary measurement science, as it allows for the quantitative visualization of samples that range from medicine to the environment. This Collection includes five papers showing the diversity of samples that can be analyzed with mass spectrometry imaging. For instance, work in the Ruman lab details the use of mass spectrometry imaging to analyze human fingerprints for forensic analysis (DOI: 10.1021/acsmeasuresciau.1c00020). This analysis provided chemical information from amino acids, fatty acids, lipids, and inorganic salts in the fingerprints that allow for the study of ridge patterns and sweat pores visually and chemically.

The Baker group showed the advantages of mass spectrometry imaging for combining histology and lipidiomics analysis for tissue samples (DOI: 10.1021/acsmeasuresciau.1c00035). Not only is mass spectrometry imaging multidisciplinary, but it can also be combined with other measurement science tools. For instance, the Sweedler group showed that combining optical microscopy, Raman spectroscopy, and mass spectrometry imaging allows for the study of microfluidic generated droplets (DOI: 10.1021/acsmeasuresciau.1c00017). Another example is the work of the Sombers group that combined mass spectrometry imaging with electroanalysis in living rats (10.1021/acsmeasuresciau.1c00030). These combined techniques really show the future of in situ and in vivo measurement science techniques.

Finally, as artificial intelligence (AI) and machine learning (ML) are taking over the measurement science field, there have been major advances in their use for complex mass spectrometry imaging as shown in a recent review article by Guarav Chopra et al. (DOI: 10.1021/acsmeasuresciau.3c00060).

Shelley D. Minteer, Editor-in-Chief, *ACS Measurement Science Au*

ANALYTICAL CHEMISTRY

It is exciting to see all of the emerging capabilities in the field of mass spectrometry imaging (MSI). Historically, MSI has its roots in SIMS and other approaches that have been used to probe the localization of specific compounds on surfaces for more than 50 years. Recent advances have included improved mass resolution, development of combined technologies such as ion mobility MALDI MS, and increases in sensitivity with MALDI 2 and enhanced matrices and derivatization schemes. There has also been significant progress in the application of multimodal approaches, proteoform imaging via nanoDESI, single-cell measurements, and construction of three-dimensional atlases, to name just a few. These advances can be related to many distinct aspects of measurement science, including enhanced sampling and sample preparation, the mass spectrometry measurement itself, data workup, and informatics.

Analytical Chemistry welcomes manuscripts that describe new chemical measurements with improved figures of merit, which often result from the addition of new dimensions of information. We publish advances almost every month; if a new technology is demonstrated, along with an enhanced application (e.g., a new sample type), this work is of interest to us. Alternatively, if your work applied an already published/established approach, such as MSI to investigate a distinct tissue type, even if method optimization was involved, the manuscript may be better suited to one of our other measurement science journals. In other words, exciting MSI work applying established protocols and instrumentation is best directed elsewhere. We also welcome ideas for Perspective

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or Review articles related to MSI, preferably via a presubmission email to the editor. Your inquiry should describe the topic, why you are the one to write the article, and any other details you think are important. While most Perspectives are invited, they can be considered without invitation. Reviews are considered invited articles that cover a specific topic; by contacting us before submission, we can determine how your idea fits with those already in our pipeline. Occasionally, author-suggested topics for Reviews are considered for publication (and an example can be seen in this Collection).

In terms of the current Collection, it was hard to choose only five articles given how many exciting MSI advances *Analytical Chemistry* has published, and so we selected those representing distinct areas of importance to MSI. We selected an exciting review highlighting recent advances in the field of MSI (DOI: 10.1021/acs.analchem.4c05249). We include manuscripts that (1) demonstrate the combination of quantitative MSI followed by LC-MS/MS proteomics and lipidomics from the same tissue area (DOI: 10.1021/acs.analchem.3c05850); (2) highlight exciting advances that enable the visualization of subcellular heterogeneity using DESI MS (DOI: 10.1021/acs.analchem.5c00843); (3) enable isobar and isomer imaging with nanoDESI (DOI: 10.1021/acs.analchem.3c04705); and finally (4) provide network analyses combined with MSI to understand molecular mechanisms (DOI: 10.1021/acs.analchem.4c06682). These five manuscripts are just a small sampling of the exciting new approaches published in *Analytical Chemistry* over the past two years. We hope you enjoy these five manuscripts and the many more we have published that demonstrate the many recent advances to MSI. Of course, stay tuned for further enhancements to MSI in the coming years published in *Analytical Chemistry* and the rest of the ACS measurement science portfolio!

Jonathan V. Sweedler, Editor-in-Chief, *Analytical Chemistry*

CHEMICAL & BIOMEDICAL IMAGING

Chemical & Biomedical Imaging (CBMI) is dedicated to publishing cutting-edge and innovative research in imaging across all fields of chemistry, physics, biology, engineering, medicine and life science (DOI: 10.1021/cbmi.4c00043). Among emerging techniques, mass spectrometry imaging (MSI) stands out for its ability to map molecular distributions across tissue samples in a label-free manner and avoiding spectral overlap, allowing simultaneous visualization of thousands of ions for detailed molecular insights. In a recent Perspective, Xu and colleagues review next-generation MSI, highlighting high-resolution methods like DESI, MALDI, and SIMS, as well as novel approaches like near-field and extreme ultraviolet laser desorption (DOI: 10.1021/cbmi.3c00061). They discuss strengths and limitations and envision future directions, including matrix-free analysis, subcellular spatial resolution, and integration with machine learning for single-cell spatial omics, emphasizing MSI's transformative potential.

MSI also advances our understanding of the behavior of nanoparticles and radionuclides within biological systems. Terry et al. used laser ablation-inductively coupled plasma-mass spectroscopy (LA-ICP-MS) with ion beam analysis to image intracellular distribution of thallium-201 (^{201}Tl), revealing its selective nuclear uptake, which impacts microdosimetry in targeted radionuclide therapy (DOI: 10.1021/cbmi.5c00113). Nuclear localization increased toxicity, while cytoplasmic localization reduced toxicity. Similarly, LA-ICP-MS studies of gold nanoparticles (AuNPs) in mice showed that positively charged AuNPs are rapidly excreted via liver and kidneys, highlighting surface chemistry's influence on distribution and clearance (DOI: 10.1021/cbmi.4c00082). These examples demonstrate MSI's critical role in tracking the fate of materials at cellular and suborgan levels for biomedical optimization.

MSI also serves as an integrative modality for spatially resolved molecular profiling in complex tissues, offering insights into both metabolic and proteomic heterogeneity. In triple-negative breast cancer (TNBC), Gartia et al. combined Raman spectroscopy with liquid chromatography−mass spectrometry (LC−MS) for multiomics, revealing changes in lipid composition and extracellular matrix (ECM) proteins, including elevated ω-3 and ω-6 fatty acids and differential collagens and fibronectin expression (DOI: 10.1021/cbmi.5c00104). These insights highlight coordinated lipid metabolism and ECM remodeling that underpin tumor progression, demonstrating the value of nondestructive spatial profiling for intratumor heterogeneity and microenvironment analysis. Complementing these advances, tyramide-amplified multimodal probes, such as a TyrRu ruthenium complex developed by Bishop et al., enhanced MSI sensitivity (DOI: 10.1021/cbmi.6c00007). Pairing tyramide signal amplification with elemental protein labeling and correlative immunofluorescence allows multiplexed analyses with high signal-to-noise ratios. Together, these approaches demonstrate the growing power of MSI to link molecular specificity with spatial resolution, facilitating detailed in situ mapping of lipids, proteins, and other biomolecules. Combining label-free spectroscopic methods with amplified elemental imaging promises unprecedented opportunities to dissect tumor heterogeneity, elucidate microenvironmental interactions, and guide precision oncology.

Zijian Guo, Editor-in-Chief, *Chemical & Biomedical Imaging*

JOURNAL OF THE AMERICAN SOCIETY FOR MASS SPECTROMETRY

New developments in mass spectrometry imaging (MSI) have expanded its versatility and scope of applications, enabling tremendous insights based on the ability of MSI to directly profile molecules in complex samples. Five recent papers published in *JASMS* spotlight innovative mass spectrometry methods and high impact applications. Because MSI often entails monitoring hundreds or thousands of chemical species for thousands of pixels per tissue section, acquisition of simultaneous MS/MS data for all species is impractical, restricting confident annotation of all chemical features. To expand the information content of nontargeted imaging, collision cross sections derived from ion mobility measurements have been used as a means to enhance identification of compounds and differentiation of isomers, as reported in the first two articles selected for this Collection. Fernandez and coworkers coupled desorption electrospray ionization with collision cross sections and machine learning predictions to increase assignment of lipids in human renal cell carcinoma tissues for an integrated spatial metabolomics workflow (DOI: 10.1021/jasms.5c00090). Zeng et al. used ion mobility with MALDI-MSI to improve differentiation and spatial profiling of disaccharide isomers, particularly sucrose and trehalose and their 6-phosphate derivatives, in poplar root and soybean root nodule tissue (DOI: 10.1021/jasms.5c00101).

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Expanding the chemical range of MSI remains another significant goal, particularly for molecules that have low abundance or poor ionization efficiencies. Work by Lim and co-workers describes the development of click-chemistry based photocleavable mass-tagged probes for multiplexed MALDI MSI of low abundance oligonucleotides in Alzheimer's mouse brain tissue (DOI: 10.1021/jasms.5c00057). The method was applied for multiomic imaging, achieving detection of both label-free metabolites (lipids) and targeted transcripts based on monitoring the photocleaved mass tag reporters. Other classes of molecules, such as certain classes of lipids, do not ionize efficiently by MALDI, and often the analysis of these molecules is further complicated by the existence of numerous isomers and isobars. The Spraggins group tackled the challenge of low ionization efficiencies of neutral lipids in MALDI-MSI by performing isotonic metal-cation washes in the sample preparation workflow as a means of salt doping to create metal-cationized lipids (10.1021/jasms.5c00202). The production of salt adducts enhanced the ionization efficiencies and separation of lipid isobars and isomers based on ion mobility mass spectrometry. The method was demonstrated for tissues from multiple organ types, including murine brain, rabbit adrenal gland, human colon, and human kidney.

To create three-dimensional depth maps of tissue compositions, MS-imaging has typically been performed by analyzing two-dimensional sections prior to reconstruction of a 3D image. In recent work by Muddiman and co-workers, an ablation-based 3D MSI technique was developed using a high-energy burst-mode ultraviolet laser in conjunction with a chromatic confocal aberration probe with automatic *z*-axis correction to measure the depth of ablation (DOI: 10.1021/jasms.5c00224). This strategy was used to image the medicinal herb *Artemisia annua* and visualize the artemisinin metabolic pathway.

Jennifer S. Brodbelt, Editor-in-Chief, *Journal of the American Society for Mass Spectrometry*

JOURNAL OF PROTEOME RESEARCH

The *Journal of Proteome Research* seeks to publish studies that advance both the technological capabilities and biological applications of modern proteomics, particularly those enabled by mass spectrometry-based analytical platforms. We are interested in papers that expand the frontiers of proteomics through innovative methodologies, spatially resolved analyses, and integrative multiomics strategies. Spatial analyses have become an area of keen interest in proteomics and metabolomics, Studies that develop or refine analytical workflows such as MALDI mass spectrometry imaging (MSI) or Direct Electrospray Ionization (DESI) protocols capable of achieving single-cell resolution or enabling proteomic analysis of archival pathology specimens demonstrate the types of methodological advances that broaden the scope of samples and biological questions accessible to proteomics and metabolomics.

The journal is also interested in research that applies these emerging technologies to biologically meaningful systems, including studies of brain aging, disease models, and subcellular protein organization. By pushing new technologies and approaches to answer important biological questions, methods are improved and legitimized. Papers that combine complementary analytical platforms such as integrated lipidomics and proteomics approaches or spatial proteomic techniques are particularly valuable because they provide a more comprehensive view of molecular organization in tissues and cells. The journal also welcomes contributions that incorporate advanced computational and machine learning approaches to improve data quality, system performance, or interpretation of complex proteomic data sets, reflecting the growing importance of informatics in large-scale proteomics research.

Collectively, the papers published in this Collection on Mass Spectrometry Imaging illustrate the journal's interest in publishing work that not only pushes the technical boundaries of proteomic measurement but also translates these innovations into deeper biological insight. The *Journal of Proteome Research* therefore prioritizes studies that combine methodological innovation, rigorous analytical validation, and clear biological relevance, particularly in emerging areas such as spatial proteomics, subcellular proteome mapping, and computational analysis of high-dimensional mass spectrometry data.

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Notes

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